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(54) Title: OPAQUE FILMS COMPRISING ISOTACTIC POLYPROPYLENE (57) Abstract An opaque polymeric film having enhanced moisture barrier and mechanical properties. The film has a base layer which includes a blend of a high crystallinity polypropylene having an isotactic stereoregularity greater than 93 % and a cavitating agent in an amount effective to cavitate the base layer.		

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OPAQUE FILMS COMPRISING ISOTACTIC POLYPROPYLENE
BACKGROUND OF THE INVENTION

The present invention relates to an opaque polypropylene-based multilayer film structure which exhibits enhanced moisture barrier and mechanical properties.

5 Opaque polymeric films are used in many commercial applications. One particularly important application is the packaging of products such as snack foods and candy bars. Films employed in the food packaging industry are chosen and/or designed to provide characteristics necessary for proper food containment. Such characteristics include water vapor barrier properties, oxygen and gas barrier properties and flavor and aroma barrier properties.

10 Polypropylene is a polymer commonly employed in the manufacture of opaque films used in the food packaging industry. In the case of multilayer films, polypropylene is typically used in the base or core layer and is cavitated with a cavitating agent such as polybutylene terephthalate (PBT). Typically, it has been necessary to use from 6% to 12% by weight of the cavitating agent in the prior art films to achieve the desired degree of cavitation.

15 However, the use of the PBT cavitating agent within the aforementioned ranges is not without its drawbacks. Particularly, the loading ranges typically employed in the art may negatively impact production uptime. Moreover, the PBT itself, at the loading ranges typically employed in the art, represents a significant percentage of the raw material costs.

20 The mechanical properties of a polymeric film structure are another important characteristic, particularly with respect to such applications as food and candy wrappings. Films having enhanced mechanical properties facilitate handling and packaging because such films are more readily accommodated by typical industrial machinery. Attempts have been made to enhance the mechanical properties of polypropylene-based films (as measured by the stiffness and moduli of the film) through increased orientation and/or by the addition of
25 additives. However, increased orientation often increases the likelihood of film splitting during manufacturing, while the addition of additives typically provides limited enhancement of mechanical properties but can negatively impact other film characteristics such as dimensional stability.

Thus, there is a need in the art for an opaque polypropylene-based film which may be readily manufactured (i.e., improved production uptime and reduced raw material costs), and which exhibits enhanced moisture barrier and mechanical properties.

The present invention, which addresses the needs of the prior art, relates to an opaque
5 polymeric film having enhanced moisture barrier and mechanical properties. The film has a base layer which includes a blend of a first polypropylene polymer having an isotactic stereoregularity greater than 93% and a cavitating agent in an amount effective to cavitate the base layer.

In one preferred embodiment, the base layer includes less than 6% by weight of
10 cavitating agent, and preferably from 2% to 5% of cavitating agent. In another preferred embodiment, the base layer further includes a resin modifier in an amount up to 9% by weight of said base layer. The resin modifier is preferably a hydrogenated hydrocarbon.

In still another preferred embodiment, at least one tie layer is adhered to the cavitated base layer. A skin is preferably adhered to the other side of the tie layer. The tie layer, which
15 facilitates adherence of the skin layer to the cavitated base layer, is preferably formed from polypropylene. The skin layer is preferably formed from an ethylene-propylene random copolymer or an ethylene-propylene-butene-1 terpolymer.

As a result, the present invention provides an opaque polypropylene-based film structure exhibiting enhanced moisture barrier and mechanical properties. These improved
20 properties are obtained without a negative impact on such film characteristics as dimensional stability. More to the point, these improved properties are obtained while using a significantly lower level of cavitating agent, thus providing improved production uptime and reduced raw material costs. The film structure further exhibits a high degree of machinability and processability.

25 The present invention is prepared by blending high crystallinity polypropylene (HCPP) with a cavitating agent in an amount effective to cavitate the HCPP. The HCPP has an isotactic stereoregularity of greater than 93%, preferably from 94% to 98%. The base layer preferably includes less than 6% by weight of the cavitating agent.

Commercially suitable PPs include Fina 3371 (available from Fina Oil and Chemical Co., Dallas, Texas), Exxon 4612 and Exxon 4052 (available from Exxon Chemical Co., Houston, Texas) and Amoco 6361 (available from Amoco Chemical Co., Chicago, Illinois). Commercially suitable HCPPs include Amoco 9117, Amoco 9119 and Amoco 9218 (available from Amoco Chemical Co., Chicago, Illinois), and Chisso HF5010 and Chisso XF2805 (available from Chisso Chemical Co., Ltd., Tokyo, Japan). Suitable HCPPs are also available from Solvay in Europe.

The HCPP has a high isotactic stereoregularity, resulting in higher crystallinity than conventional isotactic polypropylene, i.e., greater than 93%. (Conventional isotactic polypropylene is defined herein as having an isotactic stereoregularity of from 90% to 93%.) The HCPP thus exhibits higher stiffness, surface hardness, lower deflection at higher temperatures and better creep properties than conventional isotactic polypropylene. Further information relating to HCPP, including methods for preparation thereof, is disclosed in U.S. Patent No. 5,063,264.

For purposes of the present invention, stereoregularity can be determined by IR spectroscopy according to the procedure set out in "Integrated Infrared Band Intensity Measurement of Stereoregularity in Polypropylene," J. L. Koenig and A. VanRoggen, Journal of Applied Polymer Science, Vol. 9, pp. 359-367 (1965) and in "Chemical Microstructure of Polymer Chains," J. L. Koenig, Wiley-Interscience Publication, John Wiley and Sons, New York, Chichester, Brisbane, Toronto. Stereoregularity can also be determined by decahydronaphthalene (decalin) solubility and nuclear magnetic resonance spectroscopy (NMR).

The use of HCPP in the base layer unexpectedly lowers the amount of cavitating agent required to cavitate the film and produce the desired opacity. More particularly, the use of HCPP in the base layer lowers the amount of cavitating agent necessary to cavitate the film from the prior art levels of approximately 6% to 12% by weight, or greater, to a level in the present invention of less than 6% by weight, and preferably from 2% to 5% by weight. This reduction in loading level of the cavitating agent is significant in that it results in greater production uptime. In addition, the reduction in the amount of cavitating agent necessary to cavitate the base layer translates into a significant cost savings in raw materials.

More to the point, the blend of HCPP and cavitating agent in the base layer provides a significant improvement in the moisture barrier of the resultant film. As demonstrated in the example set forth hereinbelow, improvements in moisture barrier of up to 20% to 30% may be achieved with the film structures of the present invention. It will be recognized by those skilled in the art that this 20% to 30% improvement in moisture barrier is significant in the food packaging industry.

Preferred cavitating agents include polybutylene terephthalate (PBT), calcium carbonate (CaCO_3), polyester (PET), polyamides including nylon and other polymers and/or inorganic materials which are incompatible with HCPP.

In one preferred embodiment, the HCPP in the base layer is blended with a conventional isotactic polypropylene (PP). The base layer preferably includes an amount of PP effective to plasticize the base layer and thus increase the processability of the resultant extruded film structure. The blended base layer increases manufacturing efficiency by increasing line operability and line percentage uptime. For example, the blended base layer reduces the force necessary to stretch the film and in addition facilitates edge trimming of the extruded film. Thus, the blended base layer results in a reduction in cost associated with manufacture of the film structure.

In another preferred embodiment, a resin modifier is blended with the base layer precursor. The resin modifier is present in an amount of up to 9% by weight, and preferably from 3% to 6% by weight. The blending may be accomplished by the direct feeding of HCPP and resin modifier into the film extruder or by use of a masterbatch. One preferred method of blending the components utilizes a masterbatch formed of HCPP and resin modifier, e.g., 80% HCPP and 20% resin modifier. The masterbatch is then blended with additional HCPP, which reduces the concentration of the resin modifier to the final desired level.

As disclosed in commonly-owned copending U.S. Patent Application Serial Nos. 08/490,081 filed June 13, 1995 and 08/542,209 filed October 12, 1995, polymeric film structures formed from HCPP exhibit unexpectedly large decreases in water vapor transmission with small additions of resin modifier. The moisture barrier increases as the loading level of resin modifier is increased until a point is reached (less than 10% resin modifier) at which further increases in the loading level of the resin modifier do not provide

further substantial increases in moisture barrier. Moreover, the HCPP-based film structures also provide enhanced mechanical properties without suffering from losses in dimensional stability, as is common in prior art film structures which may require upwards of 20% resin modifier to achieve maximum moisture barrier.

5 The resin modifier is preferably a low molecular weight hydrogenated hydrocarbon which is compatible with the HCPP polymer and which provides the desired enhancement of film properties. The preferred resin modifier has a number average molecular weight less than 5000, preferably less than 2000, and more preferably from 500 to 1000. The resin modifier can be natural or synthetic and preferably has a softening point of from 60° to 180°C.

10 Particularly suitable hydrocarbons which can be subsequently hydrogenated are the hydrocarbon resins. Preferred hydrocarbon resins include among others petroleum resins, terpene resins, styrene resins and cyclopentadiene resins.

 Examples of commercially available hydrogenated hydrocarbon resins suitable for use in the present invention are those sold under the trademarks PICCOLYTE, REGALREZ and
15 REGALITE by Hercules Corporation of Delaware and under the trademark ESCOREZ by Exxon Chemical Company of Houston, Texas. The ESCOREZ resins are particularly preferred hydrogenated hydrocarbon resins.

 One particularly preferred resin modifier is referred to herein as a saturated alicyclic resin. The saturated alicyclic resins are obtained by the hydrogenation of aromatic
20 hydrocarbon resins. The aromatic resins are themselves obtained by polymerizing reactive unsaturated hydrocarbons containing, as the principal component, aromatic hydrocarbons in which the reactive double bonds are generally in side-chains. More particularly, the alicyclic resins are obtained from the aromatic resins by hydrogenating the latter until all, or almost all, of the unsaturation has disappeared, including the double bonds in the aromatic rings.

25 The saturated alicyclic resins used in the present invention have a softening point from 85° to 140°C, and preferably from 100° to 140°C, as measured by the ball and ring method. Examples of commercially available saturated alicyclic resins suitable for use in the present invention are those sold under the trademark ARKON-P by Arakawa Forest Chemical Industries, Ltd. of Japan.

In one preferred embodiment, a tie layer of an olefinic polymer is adhered to the base layer. Inasmuch as the skin layers discussed below may not adequately adhere to the cavitated base layer, a tie layer compatible with the cavitated base layer is first adhered to the base layer. A skin layer of an olefinic polymer is, in turn, adhered to the tie layer. Both the tie and skin
5 layers are preferably coextruded with the base layer. The tie layers may include a whitening agent such as TiO_2 .

Suitable olefinic polymers utilized for the skin layer(s) include i) ethylene homopolymers, ii) copolymers of ethylene and propylene, iii) copolymers of ethylene or propylene and butylene or another alpha olefin having 5 to 10 carbon atoms, iv) terpolymers
10 of ethylene, propylene and butylene or another alpha-olefin having 5 to 10 carbon atoms, and v) mixtures thereof.

Olefinic polymers which are particularly preferred for the skin layer(s) include ethylene-propylene copolymers with propylene as the main constituent and an ethylene content of 2% to 10% by weight (relative to the weight of the copolymer), propylene-butylene
15 copolymers with propylene as the main constituent and a butylene content of 0.5% to 25% by weight (relative to the weight of the copolymer), and ethylene-propylene-butene-1 terpolymers with propylene as the main constituent, 0.5% to 7% by weight of ethylene and 5% to 30% by weight of butene-1 (each time relative to the weight of the terpolymer), and mixtures of these polymers. The co- and terpolymers are preferably random polymers.

In a still further preferred embodiment, a coating is applied to the outer surface of the skin layer(s). An acrylic coating, which provides improved printability, machinability and aroma barrier characteristics, may be applied to one of the skin layers. A heat seal coating
20 such as ethylene methyl acrylate (EMA) or ethylene acrylic acid (EAA) may be applied to the other skin layer. Other suitable coatings include polyvinylidene chloride (PVDC), polyvinyl alcohol (PVOH) and low temperature heat seal coatings, as disclosed in commonly-owned
25 U.S. Patent No. 5,419,960.

In order to further improve certain properties of the resultant film, effective amounts of additives such as antiblocking agents, antistatic agents and/or slip agents may be contained in the base layer, tie layer(s) and/or skin layer(s).

Preferred antiblocking agents include silica, talc, clay, sodium aluminum silicate, and conventional inorganic anti-blocks. Other suitable antiblocking agents include inorganic additives, such as silicon dioxide, calcium carbonate, magnesium silicate, aluminum silicate, calcium phosphate, and the like, and/or incompatible organic polymers, such as polyamides, polyesters, polycarbonates and the like.

Preferred antistatic agents include alkali alkane sulfonates and essentially straight-chain, saturated aliphatic tertiary amines possessing aliphatic radicals with 10 to 20 carbon atoms and being substituted by 2-hydroxyalkyl-(C₁ to C₄) groups. Preferred amines are N,N-bis-(2-hydroxyethyl)-alkylamines having 10 to 20, preferably 12 to 18, carbon atoms in their alkyl groups. The effective amount of antistatic agent varies in the range from 0.05% to 3% by weight, relative to the weight of the layer.

Preferred slip agents include higher aliphatic acid amides, higher aliphatic acid esters, waxes, metallic soaps and silicone oils such as polydimethylsiloxane. The effective added amount of lubricant varies from 0.1% to 2% by weight.

The multilayer films of the present invention may be prepared employing commercially available systems for coextruding resins. As mentioned, the blended HCPP and cavitating agent precursor is preferably coextruded with at least one second and at least one third polymeric material, which form the tie and skin layers, respectively. The polymers can be brought to the molten state and coextruded from a conventional extruder through a flat sheet die, the melt streams being combined in an adapter prior to being extruded from the die. After leaving the die orifice, the multilayer film structure is quenched.

The film structure of the present invention is preferably biaxially oriented. In one preferred embodiment, the film structure is stretched from 4.5 to 6 times in the machine direction (MD) and from 6 to 13 times in the transverse direction (TD). The overall orientation (MD x TD) preferably ranges from 25 to 80. After orientation, the edges of the film can be trimmed and the film wound onto a core.

The film structures of the present invention also exhibit unexpectedly increased stiffness and moduli (MD and TD) over film structures having conventional isotactic polypropylene-based core layers. The increased stiffness and moduli provide the film structure with enhanced mechanical properties which, in turn, facilitate subsequent handling and

packaging. Moreover, the increases in MD and TD moduli are accomplished at relatively low orientation, thus reducing manufacturing costs and the associated likelihood of splitting.

The film structures of the present invention are formed having a thickness ranging from 10 microns to 60 microns, preferably from 15 microns to 50 microns. Each tie layer
5 represents from 5% to 15% of the structure, while each skin layer represents from 2% to 6% of the structure.

EXAMPLES

Water Vapor Transmission Rate (WVTR) in each of the following examples was measured at 37.8°C (100°F) and 90% Relative Humidity (ASTM F 372) and is expressed in
10 g/100 in²/day/mil.

EXAMPLE 1

Samples 1a to 1c were produced to compare the moisture barrier of an opaque PP-based film structure with an opaque HCPP-based film structure and an opaque HCPP-based film structure modified with a hydrogenated hydrocarbon.

15 Sample 1a was produced with a core layer of 94% standard PP (Exxon 4252) and 6% cavitating agent (Celanese 1300a) having a thickness of 28.25 microns, and was coextruded with skin layers of standard PP (Exxon 4252) each having a thickness of 6.0 microns to produce an ABA extrudate. The ABA extrudate was stretched 5.3 times in the machine direction and 9.0 times in the transverse direction.

20 Sample 1b was produced with a core layer of 96% HCPP (Amoco 9117) and 4% cavitating agent (Celanese 1300a) having a thickness of 26.75 microns, and was coextruded with skin layers of HCPP (Amoco 9117) each having a thickness of 5.75 microns to produce an ABA extrudate. The ABA extrudate was stretched 5.3 times in the machine direction and 9.0 times in the transverse direction.

25 Sample 1c was produced with a core layer of 93% HCPP (Amoco 9117) and 4% cavitating agent (Celanese 1300a) and 3% hydrogenated hydrocarbon (Arkon P-115) having a thickness of 24.5 microns, and was coextruded with skin layers of HCPP (Amoco 9117) each having a thickness of 5.25 microns to produce an ABA extrudate. The ABA extrudate was stretched 5.3 times in the machine direction and 9.0 times in the transverse direction.

Sample	Core Layer	% Cav. Agent	% Resin Modifier	WVTR (Aged)
5 1a	PP	6.0	0.0	.301
1b	HCPP	4.0	0.0	.242
1c	HCPP	4.0	3.0	.216

EXAMPLE 2

10 Samples 2a to 2d were produced by samples 2a to 2d were produced to compare the moisture barrier of an opaque PP-based film structure with opaque HCPP-based film structures.

Sample 2a was produced with a core layer of 92% standard PP (Amoco 6317) and 8% cavitating agent (Celanese 1300a) having a thickness of 25.0 microns, and was coextruded
15 with skin layers of 94% standard PP (Amoco 6317), 4% TiO₂ (Titanium Oxide), and 2% Talc each having a thickness of 5.35 microns to produce a ABA extrudate. The ABA extrudate was stretched 5.5 times in the machine direction and 8.5 times in the transverse direction.

Sample 2b was produced with a core layer of 30% standard PP (Amoco 6317), 66.8% HCPP (Chisso XA4141) and 3.2% cavitating agent (Celanese 1300a) having a thickness of
20 22.75 microns, and was coextruded with skin layers of 94% standard PP (Amoco 6317), 4% TiO₂ (Titanium Oxide), and 2% Talc each having a thickness of 4.85 microns to produce a ABA extrudate. The ABA extrudate was stretched 5.5 times in the machine direction and 8.5 time in the transverse direction.

Sample 2c was produced with a core layer of 45% standard PP (Amoco 6317), 51.8%
25 HCPP (Chisso XA4141) and 3.2% cavitating agent (Celanese 1300a) having a thickness of 22.75 microns, and was coextruded with skin layers of 94% standard PP (Amoco 6317), 4% TiO₂ (Titanium Oxide), and 2% Talc each having a thickness of 4.85 microns to produce a ABA extrudate. The ABA extrudate was stretched 5.5 times in the machine direction and 8.5 times in the transverse direction.

30 Sample 2d was produced with a core layer of 30% standard PP (Amoco 6317), 66.8% HCPP (Amoco 4141X) and 3.2% cavitating agent (Celanese 1300a) having a thickness of 22.75 microns, and was coextruded with skin layers of 94% standard PP (Amoco 6317), 4% TiO₂ (Titanium Oxide), and 2% Talc each having a thickness of 4.85 microns to produce a

ABA extrudate. The ABA extrudate was stretched 5.5 times in the machine direction and 8.5 times in the transverse direction.

Sample	Core Layer	% Cav. Agent	% PP ¹ in Core	% HCPP in Core	WVTR (Unaged)
2a	PP	8.0	92.0	0.0	0.47
2b	HCPP	3.2	30.0	66.8	0.39
2c	HCPP	3.2	45.0	51.8	0.39
2d	HCPP	3.2	30.0	66.8	0.40

¹ Total % PP includes 30.0% of reprocessed polypropylene

EXAMPLE 3

Sample 3a was produced to demonstrate the moisture barrier of an opaque HCPP-based film structure containing a low level of cavitating agent.

Sample 3a was produced with a core layer of 97% HCPP (Amoco 9218) and 3% cavitating agent (Celenese 1300a) having a optical thickness of 28 microns. This core was coextruded with skin layers of 92% HCPP (Amoco 9218) and 8% whitening agent (Schulman CTW5050) each having a thickness of 3.5 microns to produce an ABA extrudate. The ABA extrudate was stretched 5.3 times in the machine direction and 8.5 times in the transverse direction.

Sample	Core Layer	% Cav. Agent	% Resin Modifier	WVTR (Aged)
3a	HCPP	3.0	0.0	0.24

It is thus readily apparent from the data set forth above that the present invention provides an opaque film structure exhibiting significantly improved moisture barrier properties, as compared to the prior art control samples (samples 1a and 2a). The films of the present invention exhibited substantially the same degree of opacity as the prior art control samples (1a and 2a), while utilizing significantly lower levels of cavitating agent. Moreover, these film structures exhibit enhanced mechanical properties, e.g., improved stiffness and moduli and provide ease of manufacturing, e.g., increased line operability and line percentage uptime (as observed during manufacture of the present film structures).

CLAIMS:

1. An opaque polymeric film having enhanced moisture barrier and mechanical properties, comprising:
a base layer including a blend of a first polypropylene polymer having an isotactic stereoregularity of greater than 93% and a cavitating agent in an amount effective to cavitate said base layer.
2. The film according to Claim 1, wherein said first polypropylene has an isotactic stereoregularity of from 94% to 98%.
3. The film according to Claim 1 or 2, wherein said base layer includes up to 6% by weight of said cavitating agent.
4. The film according to Claim 3, wherein said base layer includes from 2% to 5% of said cavitating agent.
5. The film according to Claim 1 or 2, wherein said cavitating agent is selected from the group consisting of polybutylene terephthalate, calcium carbonate, polyester and polyamide.
6. The film according to Claim 1 or 2, wherein said base layer further includes a resin modifier in an amount up to 9% by weight of said base layer.
7. The film according to Claim 6, wherein said base layer includes from 3% to 6% of said resin modifier.
8. The film according to Claim 6, wherein said resin modifier is a hydrogenated hydrocarbon.
9. The film according to Claim 8, wherein said hydrogenated hydrocarbon is a hydrogenated hydrocarbon resin.
10. The film according to Claim 1 or 2, wherein said base layer further comprises an additive selected from the group consisting of antiblocking agents, antistatic agents and slip agents.
11. The film according to Claim 1 or 2, further comprising at least one tie and at least one skin layer, and wherein said tie layer is interposed between said cavitated base layer and said skin layer and is formed from a polymeric material compatible with both said cavitated base layer and said skin layer whereby said layers may be adhered together.

12. The film according to Claim 11, wherein said tie layer is polypropylene.
13. The film according to Claim 11, wherein said skin layer is formed from a polymer selected from the group consisting of ethylene-propylene random copolymers and ethylene-propylene-butene-1 terpolymers.
14. The film according to Claim 11, wherein said skin layer further comprises an additive selected from the group consisting of antiblocking agents, antistatic agents and slip agents.
15. The film according to Claim 11, further comprising a coating layer on the outer surface of said skin layer.
16. The film according to Claim 15, wherein said coating layer is selected from the group consisting of acrylic, ethylene methyl acrylate, ethylene acrylic acid, polyvinylidene chloride and polyvinyl alcohol.
17. The film according to Claim 1 or 2, wherein said blend further comprises a second polypropylene polymer having an isotactic stereoregularity of from 90% to 93%.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US97/12995

A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) : B32B 5/18, 27/32

US CL : 428/308.4, 497, 515, 516, 520; 525/55, 240; 526/348.1

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 428/308.4, 497, 515, 516, 520; 525/55, 240; 526/348.1

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
NONE

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

APS: (ISOTACTIC (P) POLYPROPYLENE), MOISTURE BARRIER?, HYDROCARBON RESIN, (OPAQUE?, VOID?, SYNTHETIC PAPER?)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 4,377,616 A (ASHCRAFT et al.) 22 March 1983, see the entire document especially column 3, lines 24-33; column 4, lines 24-39;	1,3-5, 10, and 17
Y	column 5, lines 17-21; and example 1 using "Isotactic Polypropylene", wherein the applicant himself admits that conventional polypropylene can have an isotacticity of up to 93% (page 3, line 9-10).	2, 6-9, and 11-16
Y	US 5,500,282 A (HEFFELINGER et al.) 19 March 1996, see the entire document especially column 2, lines 40+.	2, 6-9, and 11-16
Y	US 5,441,807 A (BRANT et al.) 15 August 1995, see the entire document especially column 1, lines 58+.	2, 6-9, and 1-16

☒ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:	*T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
A document defining the general state of the art which is not considered to be of particular relevance	*X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
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P document published prior to the international filing date but later than the priority date claimed	

Date of the actual completion of the international search

20 OCTOBER 1997

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24 NOV 1997

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INTERNATIONAL SEARCH REPORT

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C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 4,632,869 A (PARK et al.) 30 December 1986, see example 1.	1-17
A	US 5,147,936 A (PESZKIN et al.) 15 September 1992, see table 1 and column 3, lines 38-53 which discusses isotactic polypropylene.	1-17